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RADIOCHEMICAL ANALYSIS OF INDIVIDUAL FALLOUT PARTICLES

Research and Development Technical Report USNRDL-TR-386

17 September 1958

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U.S. NAVAL RADIOLOGICAL DEFENSE LABORATORY
SAN FRANCISCO 24 · CALIFORNIA

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NS 081-001

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Effects of Atomic Weapons

Technical Objective
AW-7

Analytical and Standards Branch
P. E. Zigman, Head

Chemical Technology Division
E. R. Tompkins, Head

Scientific Director
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Commanding Officer and Director
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San Francisco 24, California

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ABSTRACT

Quantitative measurements were made of the radioactivity of individual fallout particles from a nuclear detonation at the Eniwetok Proving Grounds. These measurements were possible since individual particles which represented approximately 10^{10} or more fissions were obtained. Although several types of particles were observed, the data were generally resolved as being derived from two major particle classes depending upon whether the coral had undergone an obvious physical alteration such as melting.

A number of individual particles were radiochemically analyzed for the nuclides Mo^{99}, Ba^{140}-La^{140}, Sr^{89} and Np^{239}. The data obtained, together with gamma spectral and decay measurements, indicate that fractionation of radionuclides was prominent in the fallout particles. Measured R values for Ba^{140} and Sr^{89} based on Mo^{99} were over an order of magnitude lower in the altered particles than in the more normal-appearing or unaltered particles. The fissions/gram values of altered particles averaged 100 times that of unaltered particles. Gamma decay curves of the two classes of particles taken from H+50 to H+10,000 hr showed marked dissimilarities.

In addition to the established feasibility of the individual particle measurements it is postulated that the radioactive composition of fallout at any point may be determined by the relative numbers of the two major classes of particles observed.

SUMMARY

The Problem

With few exceptions all studies to define the radiochemical composition of fallout have been conducted with total fallout collections, i.e., with fallout collections each of which was composed of many individual particles. The general application of such data to hazard definition and prediction and to such problems as fallout formation theory, radiological decontamination, fractionation of radioactive nuclides, etc. has been restricted by a lack of knowledge concerning the nuclide composition of individual fallout particles.

The purpose of the present work was to determine the feasibility of individual particle analysis. Feasibility was defined as the ability to quantitatively assess individual particle radioactivity. It was estimated that individual particles containing radioactivity from approximately 10^{10} fissions would be required for such assessment. It was further intended that if, indeed, such analyses were feasible, sufficient data would be obtained to establish amounts of several nuclides and to search for differences (i.e., fractionation) in radionuclide composition between individual particles.

Findings

Measurements were made of the radioactivity of approximately 1000 fallout particles from a nuclear detonation of Operation REDWING. Feasibility of individual particle analysis methods was established by radiochemical analysis. Differences in radioactive composition were observed between normal-appearing coral particles and particles which had undergone obvious physical alteration such as melting.

ADMINISTRATIVE INFORMATION

The work reported herein was an outgrowth of Project 2.63 of Operation REDWING. This research was sponsored by the Bureau of Ships of the U.S. Navy under NS 081-001 (Technical Objective AW-7) as part of Problem 3, Program 1, during FY 1957.

ACKNOWLEDGMENTS

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The assistance of Mr. H.J. Huckolls and Mr. L.J. Graham in various phases of the work is gratefully acknowledged.

CONTENTS

LIST OF FIGURES

LIST OF TABLES

INTRODUCTION

The radiochemical compositions of fallout from several nuclear weapons tests have been studied in many investigations.[1-7] With few exceptions all such studies were conducted with large fallout collections, i.e., with fallout collections each of which was composed of many individual particles. Although fallout particles have been examined on an individual basis,[8-17] very few investigations of their radiochemical compositions have been reported.[8-11] Moreover, the radioactivity data obtained in these particle studies were of a relative nature.

The considerable amount of radiochemical work on total mixtures and the work on individual particles has been useful to define fallout hazards and to develop theories for prediction of such hazards. However, it may be stated without elaboration that the general application of such data to hazard definition and prediction, and to such problems as fallout formation theory, radiological decontamination and fractionation of radioactive nuclides, has been restricted by a lack of knowledge concerning the nuclide composition of individual particles. The acquisition of such knowledge by radiochemical analysis of single fallout particles is a logical extension of both past and present studies on gross samples of radioactive fallout. Such analyses have the advantage of starting with simple systems from which theoretical deductions may be derived for more complex systems.

Since little quantitative information was available concerning levels of radioactivity or of specific nuclides associated with single particles of fallout the primary purpose of the present work was to explore the feasibility of such analyses. For the purposes of this investigation feasibility was defined as the ability to quantitatively assess individual particle radioactivity. It was estimated that individual particles containing radioactivity from approximately 10^{10} fissions would be required for such assessment. It was further intended that, if, indeed, such analyses were possible, sufficient data would be obtained to establish amounts of several nuclides and to search for differences (i.e., fractionation) in radionuclide composition between individual particles.

Measurements were carried out on particles from a nuclear detonation of Operation [redacted].[18] Individual particles were examined to determine total radioactivity and to estimate the number of fission events leading to such radioactivity. Analyses were made of the Sr^{89}, Mo^{99}, Ba^{140} and Np^{239} content and of the weight of separate particles. Gamma ray spectra and rates of decay of particles were obtained.

PROCEDURES

This Section considers three topics. These are counting instrumentation, calibrations and measurements.

Description of Counting Instrumentation

Several types of counting instruments were used in these experiments. Brief descriptions of these instruments are given below.

Gamma well counter

The gamma well counter (termed the "WC") consists of a cylindrical well-type NaI(Tl) crystal, 1-3/4 in. in diam. by 2 in. long, coupled to a 6292 DuMont phototube which in turn is connected to a preamplified and scaling circuit. The unit is capable of high count rates. Linear response up to 1,000,000 cpm is obtained with high-purity, fast decaying calibration standards. The counting efficiency of the system increases with decreasing energy of the gamma ray under assay (except at very low energies).

High-pressure gamma ionization chamber

The high-pressure gamma ionization chamber (termed the "GIC") is similar in design and operation to the chamber described by Jones and Overman.[19] This instrument provides a near 4-pi assay geometry by means of a 1-3/4 in. diam. by 8-1/2 in. long thimble which extends into a cylindrical steel chamber. The chamber contains argon gas at 600 psig and a collecting electrode consisting of a wire mesh grid surrounding the thimble. The chamber is connected externally to variable resistors and to a vibrating reed electrometer. All data procured on this instrument were normalized relative to a 100 µg radium standard response of 560×10^{-9} ma of ionization current. Although the counting efficiency of the instrument is low, it is useful for the assay of a wide range of radioactivity (for example, the instrument is sensitive to 0.02 to 1000 µc of Co^{60}). The counting efficiency of this instrument is a function of gamma ray energy and increases with increasing gamma energy.[20]

Proportional beta counters

The scaling portions of these counters are of conventional nature. Detectors consist of semi-cylindrical longitudinal chambers containing 1-mil tungsten wires. The counter gas is 90 % argon and 10 % carbon dioxide by volume. The gas flows continuously above a 1 mg/cm^2 aluminized Mylar window. Solid counting samples are contained in filter paper discs mounted on brass planchets which are indexed to a reproducible position in aluminum holders. Liquid samples are counted after placement on thin plastic films stretched over holes in aluminum cards.

Single-channel gamma pulse height analyzer

The unit consists of a 1-1/2 x 1-in. NaI detector, linear amplifier, analyzer, step scanner, and digital recorder. The small crystal employed with the analyzer is useful for scanning the low energy portion of the fission product spectrum for photopeaks from Np^{239} and Tc^{99}.

Calibrations

Prior to receipt of the fallout samples a number of preliminary experiments were made using enriched uranium (93.2 % U^{235}) samples irradiated in a high neutron flux for periods less than 30 min. The purpose of these experiments was to investigate the accuracy and reproducibility of individual measurements which were contemplated, establish a procedure for manipulations of the fallout samples, define minimum acceptable activity levels, inter-calibrate counting instruments, and provide "normal" decay curves for radioactive mixtures produced by thermal neutron fission of U^{235}.

Measurements

Three primary types of analytical determinations were carried out. These were radioactivity measurements, radiochemical analyses and weight determinations. The specific determinations and the reasons for the determinations are described below.

Radioactivity measurements

Most of the activity determinations consisted of measurement of gamma radiation with either (or both) the GIC or the WC. The values obtained on these instruments were used for primary assay purposes, to establish rates of decay and to compare the gamma activities of different particles by calculation of three activity ratios. These ratios were: (a) well counts per minute per milliampere (WC cpm/ma); well counts per minute per 10^4 fissions (WC cpm/10^4f); and milliamperes per 10^4 fissions (ma/10^4f).

Both the numerator and the denominator values of the ratio WC cpm/ma were obtained with instruments whose detection efficiencies varied with gamma energy. As noted above, the efficiency of the well counter decreased with increasing gamma energy, whereas the efficiency of the ionization chamber increased with increasing gamma energy. Consequently, the ratios WC cpm/ma for single particles (at a given time) reflect differences in the radionuclide compositions of the particles. Thus, the WC cpm/ma ratios obtained at the same time for two particles may be compared to investigate gross differences in radionuclide compositions of the two particles. In addition, the WC cpm/ma ratios of a large number of samples have the advantage of being easily measured without recourse to fission determinations.

Computations of the ratios WC cpm/10^4f and ma/10^4f were carried out for the following reasons. In the examination of radioactive fallout it is common practice to present analytical results in terms of fission events which the sample radioactivity (or sample radionuclide composition) represents, that is, the number of fission events which result in the sample radioactivity (or sample radionuclide composition). The value for number of fission events is determined by radiochemical analysis for Mo^{99} as described later in this report. Using fissions (i.e., 10^4f*) as the denominators in the ratios has the advantage of providing a single comparison base which, in itself, is not dependent upon time of analysis and which does not change with time. Moreover, each ratio reflects radionuclide composition. As such, it is possible to compare either the WC cpm per fission or ma per fission ratios of a number of particles to distinguish differences in radionuclide compositions of the particles. In addition, the ratios may be used to compute total fissions for particles which have not been subjected to radiochemical analysis but have been assayed on the well counter and ionization chamber.

Other radioactivity measurements were carried out with the single channel pulse height analyzer and with the beta counter. The analyzer was used to measure Mo^{99} and Np^{239} (as described below) and also to obtain gamma spectra which were employed in the interpretation of other measurements. The beta counter was used in the assay of individual separated fission products.

Radiochemical analyses

A number of individual particles were dissolved and radiochemically analyzed to investigate differences in radionuclide composition between the particles. The particles were placed in a small volumetric flask under a low-power binocular microscope. The sample was then dissolved by

*The unit "10^4 fissions" is widely used as a convenient computational base at this laboratory for radiochemical and radioactivity measurements.[21,22]

application of various reagents. Most of the solid material (and activity) of a single particle dissolved in a few drops of 6M HCl. The remaining solids usually consisted of small black particles which appeared to be carbonaceous and which were brought into solution by the addition of one or two drops of 72 % $HClO_4$ and warming.

Radiochemical measurements of Sr^{89}, Mo^{99}, Ba^{140}-La^{140} and Np^{239} were carried out. Each of these specific nuclides was selected because it satisfied one or more of the following criteria:

a. The nuclide was considered a reference nuclide or nuclide of particular interest from previous work (e.g., Mo^{99} and Np^{239}).

b. The nuclide was known to disproportionate (i.e. fractionate) in its distribution in fallout. For example, Sr^{89} has been found to fractionate severely between samples from the same detonation.[3,4]

c. The half-life, decay scheme and instrument response factors of the nuclide were well known. It was desired that the nuclide decay to a stable daughter and that the nuclide constitute the sole or major radionuclide of the element present at time of separation.

d. The nuclide contributes 1 % or more to the total fission product beta disintegration rate at some time during the arbitrary time period of 1 to 28 days post-detonation.

e. Well established and reasonably rapid radiochemical procedures were available for the nuclide.

Conventional radiochemical procedures were used in the analyses of Sr^{89}, Ba^{140}-La^{140}, and, in some cases, Mo^{99}.[23,24] Normally aliquots of a single solution of the dissolved sample were used. However, with low activity samples it was necessary to employ a sequential analysis of the entire sample solution. Some Mo^{99} analyses were carried out by use of the single-channel pulse height analyzer. The latter technique is based on analysis of the photopeak area of daughter Tc^{99m}.[25] The agreement between the two methods was usually within 10 % when carried out on the same samples. Similar photopeak area analyses were used to determine amounts of Np^{239}.

The radiochemical results were calculated in terms of Np^{239} product-to-fission (p/f) ratios or R values. The former values were calculated directly from the numbers of atoms of the induced activity Np^{239}, and the total fissions as calculated from the number of atoms of Mo^{99} for

each sample. The R values for Ba^{140} and Sr^{89} were defined in the usual manner by the ratio of atoms of the nuclide to the atoms of the reference nuclide, Mo^{99}, as observed in the sample divided by the same ratio for thermal neutron fission of U^{235}. From this definition it is apparent that R values from fallout samples indicate the combined effects of fractionation and variations in fission yield. Although lower than thermal fission yields for Sr^{89} and Ba^{140} were expected for fast neutron fission,[26,27,28] the deviation was considered to be not greater than 20 %. For Mo^{99} the deviation was probably less and therefore all fission calculations were based on a thermal fission yield for Mo^{99} of 6.1 %. These same considerations together with experimental uncertainties generally lead to observed R values of 1.0 ± 0.5, which are considered "normal" within these limits.

Weight Measurements

Selected particles were weighed. Most of these measurements were carried out on an Ainsworth microbalance. The data obtained were used to compute the values for the number of fissions per unit weight of fallout particles. Such values were compared to similar values obtained with gross collections of fallout materials.

SAMPLES

A total of 300 particles were received from the laboratory aboard the manned ship, YAG-40, which was positioned to receive fallout from the event.* The particles were obtained from greased trays exposed sequentially during fallout by a special collector arrangement. These trays had been examined under a microscope on the YAG-40 and randomly selected particles removed with individual needles. A small amount of grease had been placed on the needle before the particle was removed; this insured adhesion of the particle. Prior to use, each needle was mounted in a cork, allowing the needle and particle to be inserted in a small glass vial for shipment to USNRDL. After receipt at USNRDL, each particle was measured on the GIC and WC at approximately H+72 hr; the most active samples assayed approximately 10^5 cpm in the WC. Thirteen of the more active particles were selected for analyses. Most of these were dissolved; a few were retained intact for gamma spectral measurements and decay determinations.

Additional particles were obtained from two gross fallout samples returned to USNRDL by Project 2.63. These samples consisted of fallout collected in a large tray on New Island (designated OCC-NCW-F-67) and a WHIM sample** removed from the deck of a platform on the barge YFNB-29, which had been anchored in the lagoon.

The large tray was exposed to fallout for the period H+1/4 to H+11 hr. After return to USNRDL, 21 particles were removed at random with a small spatula and placed on glass slides for microscopic observation, and were later transferred to individual glass tubes. The tray was assayed for radioactivity in a low geometry scintillation counter[30,31] before the particles were removed. After the particles were removed, they were spread over the surface of a similar tray and were assayed in the same counter. Following this, each particle was subjected to analysis.

Particles were removed from the WHIM sample before and after sieving of the entire sample. Eighteen individual particles were removed before

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* Detailed descriptions of the ship positions during the event are included in the report of Project 2.63.[29]

** A non-scheduled sample.

sieving.* These were designated 165W through 182. Of these, two (170 and 177) were analyzed radiochemically. The remaining sixteen particles were gamma counted and weighed. The entire sample was then sieved, and each size fraction weighed, measured in the GIC, and placed in separate dishes. Under a microscope individual particles were scooped from large numbers of particles on each dish. Each particle was held under the microscope, typed, and placed in a separate vial for assay in the WC. Almost 650 particles were assayed. After this, a number of particles typed and designated WI (see below) from the 500 to 1000-μ sieve fraction were combined and treated as one sample. The same procedure was followed with a number of particles typed YS and a number typed WS.

Selection of Individual Particles

In all cases selections of individual particles from large groups of particles were made in a random manner except for the 18 particles from the WHIM sample as described earlier. Upon microscope examination a number of observers agreed that the following colors and shapes well describe the particles (abbreviating symbols used are listed):

Color		Shape	
Yellow	-Y	Spherical	-S
White	-W	Flaky	-F
Gray	-G	Irregular	-I
Colorless	-C	Angular	-A

Some particles which were comparatively light in color were observed; this lesser intensity was noted by addition of the letter "L" to the color designator. More detailed descriptions of particle classification parameters have been reported elsewhere.[32,33]

It was generally agreed that the spherical and flaky particles represented an obvious physical alteration of the normal-appearing (angular or irregular) coral particles. In some instances, it was difficult to type individual particles by color. However, particles which had obviously been physically altered in some manner were usually easily distinguished. As a consequence, the information obtained in this investigation is considered to have been derived from two general classes of particles - altered and unaltered. The altered were defined as particles possessing spherical shapes (presumably melted) or particles which were flaky; unaltered particles were defined as irregular or angular particles. Adams has stated**

* This group of 18 particles was not selected at random. Attempts were made to secure active spherical particles; however, a mixture of spherical and irregular particles were obtained.

** C.E. Adams, USNRDL, personal communication.

that almost all particles of both classes have actually undergone some chemical alteration and has presented data[32] which indicates that altered particles as here defined are simply normal coral particles which have been heated up to and above melting and boiling temperatures.

The only exception to the general classifications adopted above were a few particles typed as yellow irregular (YI) but which were easily recognized as portions of broken spheres. These were, therefore, altered particles.

RESULTS

The majority of the analytical data obtained are presented in three tables and one figure. One table includes all counting and radiochemical data for all individual particles studied. The figure shows the activities of a large number of particles selected at random from the WHIM sample. Additional data for the WHIM sample are given in the second and third tables. Data describing decay and gamma spectra are presented later in this report in the section where the counting and radiochemical results are discussed. This method of presentation was adopted since the primary purpose of the decay and spectral data was to aid in the interpretation of the counting and radiochemical results.

In the listings of data, particles are described on the basis of color and shape. A pictorial representation of several particle types is provided in Fig. 1.

Radiochemical and Counting Data for Individual Particles

Table 1 includes data for particles which were selected for individual analysis. Measurements of Ba^{140} and Sr^{89} content are expressed as R values. Well counter and GIC assays are given at times of measurement in units of counts per minute (CPM) and milliamperes of ionization current (ma), respectively. Limits of reliability are indicated by the number of significant figures and are discussed later in the report.

Physical and Counting Data for the WHIM Sample

Table 2 presents weight and activity values for the WHIM sample. The calculations for total fissions were based on empirical GIC values for ma/fission versus time observed on a gross sample of fallout collected at HOW Island and analyzed at NRDL.[29]

Counting Data for Individual Particles Selected From the Whim Sample

Data for a number of particles from the WHIM sample are presented in Table 1. Additional particles were selected from three size fractions

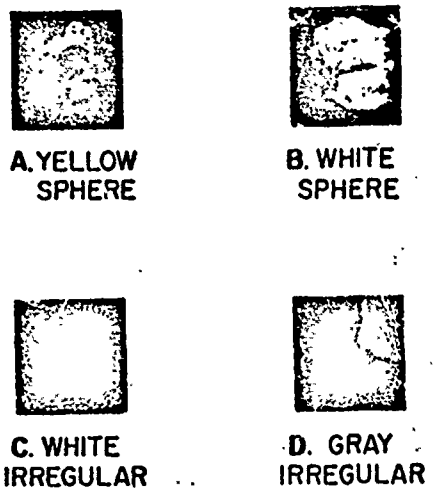

Fig. 1 Particle Types Observed in the Whim Sample. Altered particles are shown in A and B, unaltered particles in C and D.

TABLE 1

Counting and Radiochemical Results for Individual Particles

Source	Field No.	Lab. No.	Type	WC Assay		GIC Assay		Total Fissions (10^{10})	R Value		Weight (mg)
				Value (10^6 cpm)	Time (H+hr)	Value (10^{-11} ma)	Time (H+hr)		Ba^{140}	Sr^{89}	
YAG 40	331-1	1	WI	0.358	70	50	70	0.67			0.0094
	331-2	2	WS	0.0387	70	7	70				
	331-5	5	YS	0.291	70	25	70	0.72		0.015	
	331-6	6	YS	0.527	70	41	70	1.4			
	331-8	8	YI	0.255	71	26	71	0.80	0.12	0.030	
	335-2	11	WI	0.0125	71	-	-				
	335-4	13	WI	0.217	71	26	70	0.67	0.27	0.075	
	335-22	29	WI	0.0411	72	2	71				
	324-35	73	WI	0.237	73	19	73	0.49		0.15	
	324-42	80	YS	0.210	74	19	73	0.79		0.19	
	324-50	88	WI	0.266	75	29	74	0.34		0.24	
	324-58	96	WI	0.0173	75	3	74				
	325-30	128	WS	0.172	75	16	74				
OCC	How-F-67	148	GI	3.61	98	432	98	6.4	2.3	0.54	16.440
		149	I	1.71	99	183	99				6.110
		150	WI	3.52	99	172	99	1.9	2.2	1.1	8.600
		151	WI	2.48	100	316	100	3.7	3.4	0.66	8.170
		152	YI	8.80	100	497	100	22.	0.12	0.027	2.196
		153	WI	0.0361	103	3	103				3.530
		154	Metal	0.0211	103	3	103				0.303
		155	I	3.33	103	257	103	2.6	0.76	0.34	3.200
		156	WI	1.12	104	141	103	1.0	3.1	1.5	4.123
		157	YS	1.17	104	108	104				
		158	I	0.0342	104	4	104				0.171
		159	S	19.3	105	1320	105	75.	0.015	0.0025	2.246
		160	GI	0.751	105	95	105	0.64	3.4	1.5	0.920

Continued

TABLE 1 (Cont'd)

Counting and Radiochemical Results for Individual Particles

Source	Field No.	Lab. No.	Type	WC Assay Value (10^6 cpm)	WC Assay Time (H+hr)	GIC Assay Value (10^{-11} ma)	GIC Assay Time (H+hr)	Total Fissions (10^{10})	R Value Ba^{140}	R Value Sr^{89}	Weight (mg)
OCC	Hew-F 67	161	GI	0.208	105	24	105				1.883
		162	WI	0.830	105	116	105	0.87			0.872
		163	WS	6.75	119	511	119	17.	0.17	0.026	3.302
		164	WI	0.114	119	16	119				0.393
		165	WI	0.110	119	12	119				0.668
		166	GI	0.0943	119	10	119				2.738
		167	GI	0.480	119	64	119				1.672
		168	GI	0.207	119	26	119				1.137
YFNB 29	Whim (Prior to sieving)	165W	YS	17.5	239	1090.	239				6,90
		166W	YS	37.8	239	3300	239				17.3
		167W	I	3.42	239	260	239				40,1
		168W	YS	36.6	239	2720	239				8.70
		169	I	0.101	239	-	239				11,9
		170	WI	1.94	239	323	239	16.	1.5	0.42	48,3
		171	S	6.30	239	244	239				
		172	S	16.7	239	895	239				
		173	I	2.23	239	298	239				11,4
		174	S	24.3	239	1400	239				7,10
		175	S	9.05	239	490	239				2,50
		176	GI	0.444	239	51	239				48,8
		177	S	2.63	239	164	239	49.	0.026	0.0080	1,73
		178	I	1.76	239	189	239				388,
		179	YS	6.72	239	465	239				5,10
		180	I	1.71	239	199	239				457,
		181	YI	28.0	239	1720	239				25,8
		182	I	0.071	239	-					9,00
YFNB 29	Whim (After sieving)	Particles size 500-1000 μ	WI	1.07	532	226	481	44.	0.70	1.19	11,34
			YS	2.79	532	360	481	190.	0.024	0.017	2,76
			WS	1.64	532	199	481	150.	0.013	0.016	1,76

TABLE 2

Weight, Activity, and Fission Values for the Sized Fractions From the WHIM Sample

Size Range (μ)	Weight		GIC Assay			Fissions	
	Grams	Percent of Total	Value[a] (10^{-5} ma)	Percent of Total	Specific Activity[a] (10^{-5} ma/gm)	Total (10^{14})	Per Gram (10^{14})
1000	37.70	41.8	1.08	15.8	0.0286	21.	0.56
500-1000	41.91	46.4	3.14	46.0	0.0749	60.	1.4
250- 500	4.97	5.5	1.35	19.8	0.272	26.	5.2
100- 250	3.51	3.9	0.734	10.7	0.209	14.	4.0
50- 100	0.80	0.9	0.155	2.3	0.194	3.0	3.8
50	1.38	1.5	0.371	5.4	0.269	7.1	5.1
Total	90.27		6.83		0.0757	131.	1.5

a. at H+262 hr.

and were measured in the gamma well counter. A total of 639 particles were removed from the three indicated size ranges. Of this total, 211 particles were uncertain as to description and therefore were not typed. The activities of the remaining 428 particles are given in Table 3 and are shown in Fig. 2. Since decay corrections were small over the periods of measurement, the activities may be considered to be that at H+382 hr.

TABLE 3

Activity at H+382 Hr of WHIM Sample Particles From Sized Fractions

Size Range (μ)	Particle Type	Number of Particles	Total WC Activity (10^6 cpm)
500-1000	YS	11	14.5
	LYS	10	10.8
	WS	11	7.84
	WI	57	7.31
	GI	21	1.22
250-500	YS	43	16.9
	LYS	18	6.25
	WS	24	3.63
	WI	156	7.29
	GI	12	0.0913
100-250	YS	17	3.62
	LYS	4	0.450
	WS	9	0.611
	WI	33	0.277
	GI	2	0.00194

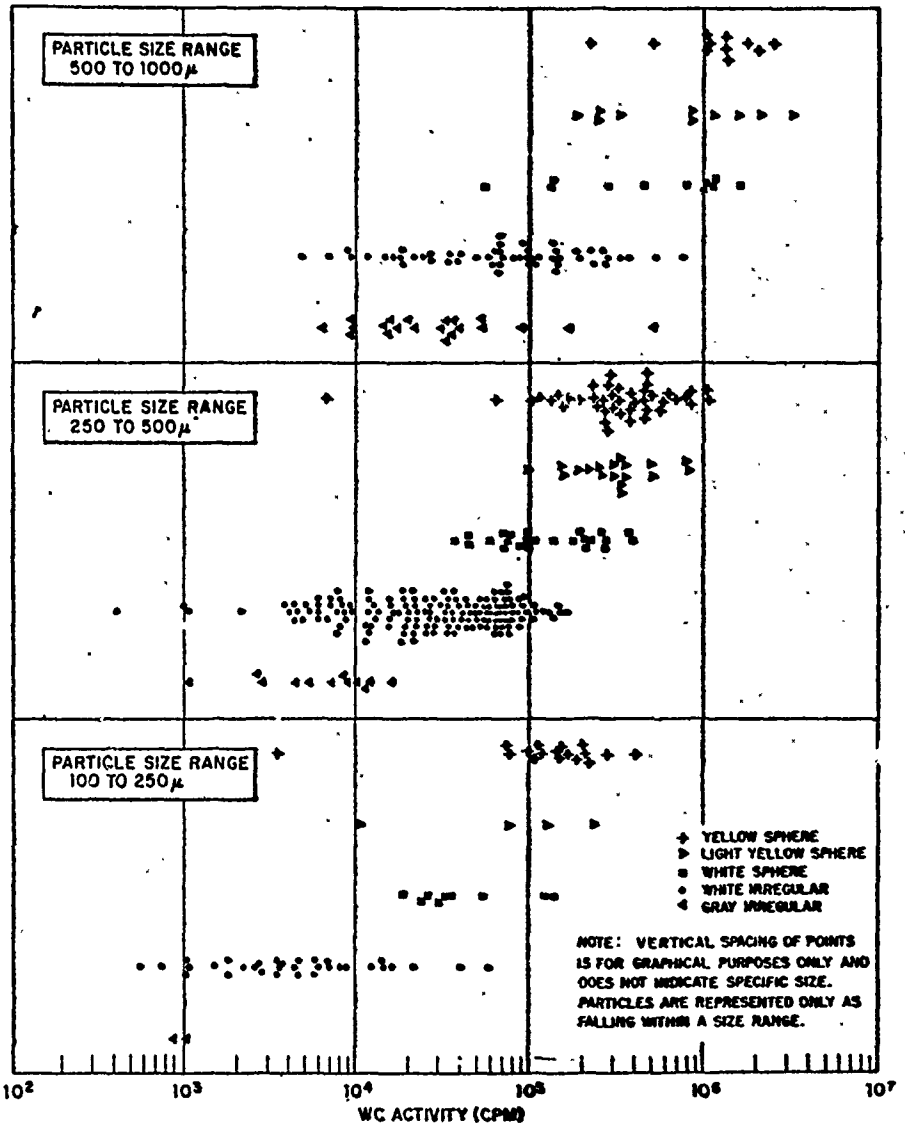

Fig. 2 WC Activity (H+382 hr) of Whim Sample Particles From Sized Fractions

DISCUSSION

Data have been presented for radiochemical analyses of a number of individual fallout particles. Analyses of these particles were possible because they contained radionuclides from approximately 10^{10} fissions (or more).

Some discussion of particle types has been presented earlier. It was stated that the coral particles could be grouped into two broad categories or classes - altered and unaltered, depending upon whether the physical appearance of the coral particles resembled a spherical or flaky (altered) shape or an angular or irregular (unaltered) shape. Division of the particles into two such broad classes was convenient in that it permitted the data to be inspected for gross differences. However, it should be realized that such divisions are not sharply bounded since differences exist between particles in a single class. Some differences were observed at this investigation. Others are implied in Williamson's[33] data, which indicates that the yellow spherical particles were slightly more radioactive and more dense than the white spheres. These latter indications were in qualitative agreement with the count data of this investigation (see Table 3 and Fig. 2) and with chemical spot tests which were made for iron and which consistently indicated greater amounts of iron in the yellow samples than in the white.

It is possible that additional types of particles existed and were not observed in all samples. For example, the YAG-40 personnel estimated that 13 % of the particles from the greased trays were of a delicate flaky or aggregated composition as opposed to the spherical and irregular particles. Flaky particles were not observed in the WHIM and OCC samples, very possibly due to the sampling methods which in some cases included sieving. Uncertainties of this nature, the physical appearance of the particles, and inspection of the data led to the concept of two broad classes of fallout particles.

In order to investigate the data for differences in radioactive composition three assumptions were made as follows:

1. Particles may be classified into two categories, physically altered or physically unaltered;
2. Particle radionuclide composition depends only on particle category. Thus, although the numbers and size ranges of the

two categories of particles may vary with time and point of collection, particles of the same category have similar radioactive compositions.

3. Fractionation of the reference nuclide Mo^{99}, is small compared to the observed differences between particle categories.

In view of these assumptions an inspection of the data in Table 1 immediately reveals differences in the radioactive composition of the two classes of particles observed. These differences are shown in Fig. 3 (A,B,C) and Table 4. The figures show the time-dependent activity ratios WC cpm/10^4f, ma/10^4f, and WC cpm/ma as a function of time of measurements for both altered and unaltered samples and also show, for comparison, similar data obtained in calibration runs with enriched U^{235} in which the Np^{239} contribution was low. Deviations of the values from the latter curve indicate the combined effects of fractionation, fission yield variations, and induced activities. The relative positions of the points clearly show the differences in the time-dependent activity ratios for the two types of particles. The mean values for these ratios at the various times of measurement are given in Table 4 along with average values for time-independent quantities which were also derived from the individual particle data in Table 1. The number of determinations and standard deviations of the mean values are also given in Table 4.

The data of Table 4 show that the value of fissions/gram was much larger in the altered particles than in the unaltered particles. The R value data indicates that the altered particles were markedly depleted in Ba^{140}-La^{140}, whereas the unaltered particles were enriched in Ba^{140}-La^{140}. Depletion of Sr^{89} occurred in altered particles and, possibly, in unaltered particles. The R values are shown graphically in Fig. 4 (A and B) in which the numbers of atoms of each nuclide are plotted versus the number of Mo^{99} atoms found in the same sample. The straight lines represent estimated fission yields for Ba^{140} and Sr^{89}. Product-to-fission (p/f) ratios for Np^{239} did not exhibit as wide a variation between particle class as either of the fission products studied. The p/f ratios obtained from individual particles were comparable to standard cloud sample values[29] but, on the average, were higher in samples of unaltered composition.

Other measurements were made on three separate groups of particles from the WHIM sample, which confirm the noted differences between altered and unaltered particles. The first group all consisted of WI (i.e., unaltered) particles; the second and third groups consisted of YS and all WS (i.e., altered particles), respectively. The data obtained is shown in Table 1. Once again, a greater number of fissions were associated with altered particles and the R values show a marked depletion of Ba^{140}-La^{140} and Sr^{89} in the altered particles.

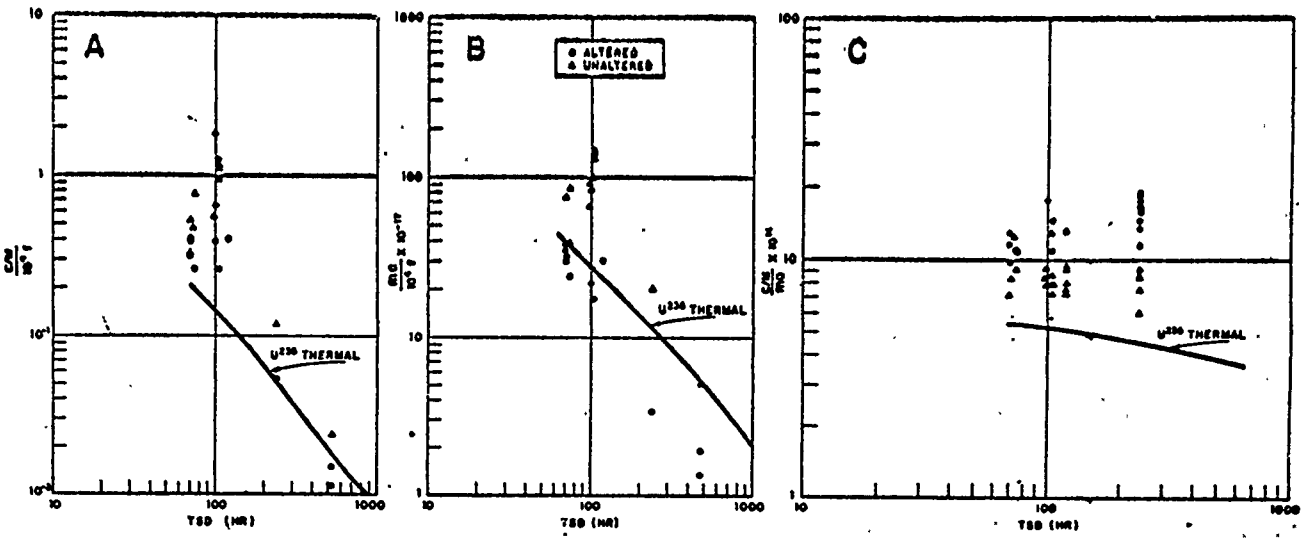

Fig. 3 Activity Ratios as a Function of Time
A. WC Activity/10^{4} Fissions.
B. GIC Activity/10^{4} Fissions.
C. WC Activity/GIC Activity.

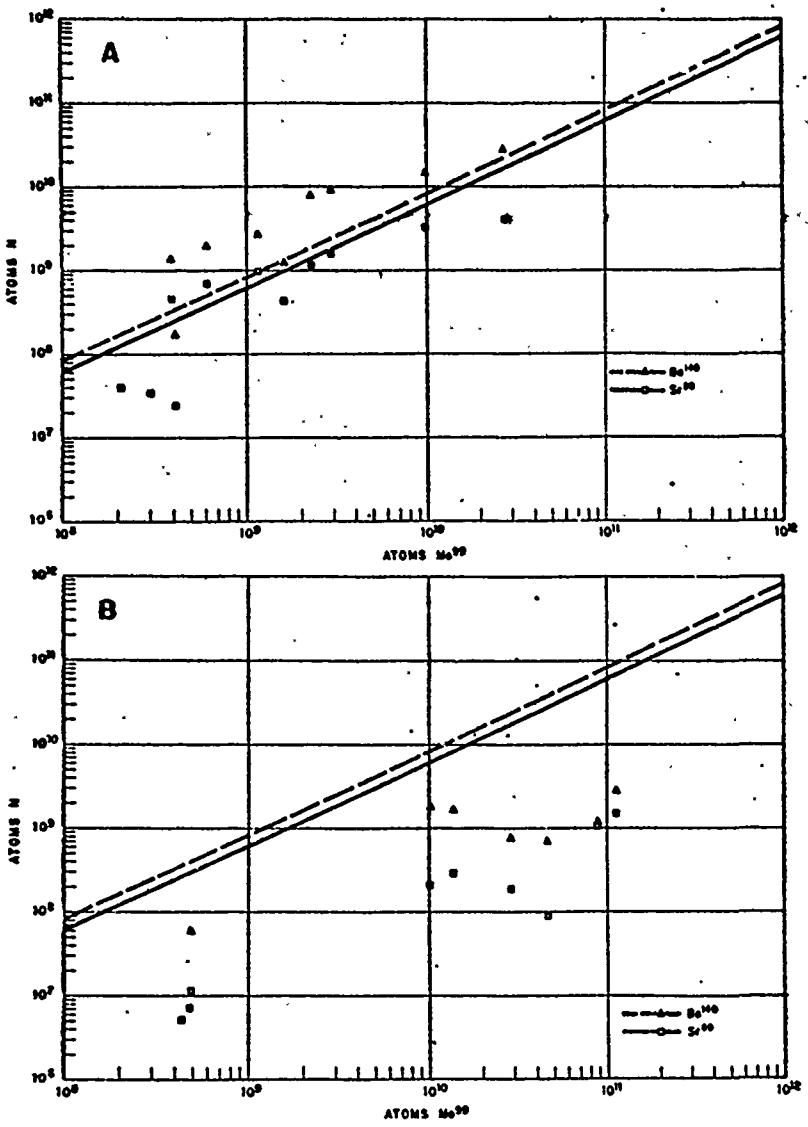

Fig. 4 Atoms of Sr^{89} and Ba^{140} Versus Atoms of Mo^{99}
A. In Unaltered Particles.
B. In Altered Particles.

TABLE 4

Mean Values for Several Quantities, for Altered and Unaltered Particles

Quantity	Time (H+hr)	Altered		Unaltered	
		No. of Samples	Value	No. of Samples	Value
fiss/gm($\times 10^{14}$)	-	6	3.8 ± 3.1	9	0.090 ± 0.12
Ba^{140}-R value	-	5	0.090 ± 0.068	8	2.1 ± 1.2
Sr^{89}-R value	-	7	0.018 ± 0.010	10	0.65 ± 0.17
WC cpm/10^4f	71	4	0.34 ± 0.06	4	0.53 ± 0.19
	105	3	0.35 ± 0.08	7	1.1 ± 0.4
	239	1	0.054	1	0.12
	532	2	0.013	1	0.024
ma/10^4f($\times 10^{-17}$)	71	4	30 ± 5	4	59 ± 24
	105	3	24 ± 7	7	109 ± 31
	239	1	3.4	1	20
	481	2	1.7	1	5.1
WC cpm/ma($\times 10^{14}$)	71	5	11 ± 1	4	9.3 ± 2.0
	105	4	14 ± 3	13	8.6 ± 1.5[a]
	239	10	16 ± 2	6	8.2 ± 1.3

a. Does not include one value, 20.5×10^{14} for sample #150.

The above data establish differences according to particle class. Although additional data would be required for quantitative assessment of these differences it was possible to study some of their effects by observation of gross decay rates, gamma ray spectra, fission per gram values, and R values.

Decay measurements

Gross decay measurements were carried out in the gamma well counter for two altered samples (No. 6 and No. 128) and two unaltered samples (No. 1 and No. 96). The curves for samples No. 1 and No. 6 are shown in Fig. 5A. The pronounced change in decay rates after 1000 hr for the altered samples as compared to the unaltered samples was verified by counts taken on a number of individual particles at H+400 and H+7100 hr. The mean ratio of the late count divided by the early count for 37 samples of the altered category was 0.015± 0.008, or approximately twice the corresponding ratio of 0.0085 ± 0.0042 found for 65 unaltered samples. These values are in good agreement with decay curve values from Fig. 5A, which give 0.015 and 0.0076 respectively for the altered (No. 6) and unaltered (No. 1) samples. The differences in decay rates are again shown in Fig. 5B, where the decay curves are plotted in terms of 10^4 fissions. Plots of the type shown in Fig. 5B, should coincide for samples with equivalent radioactive compositions.

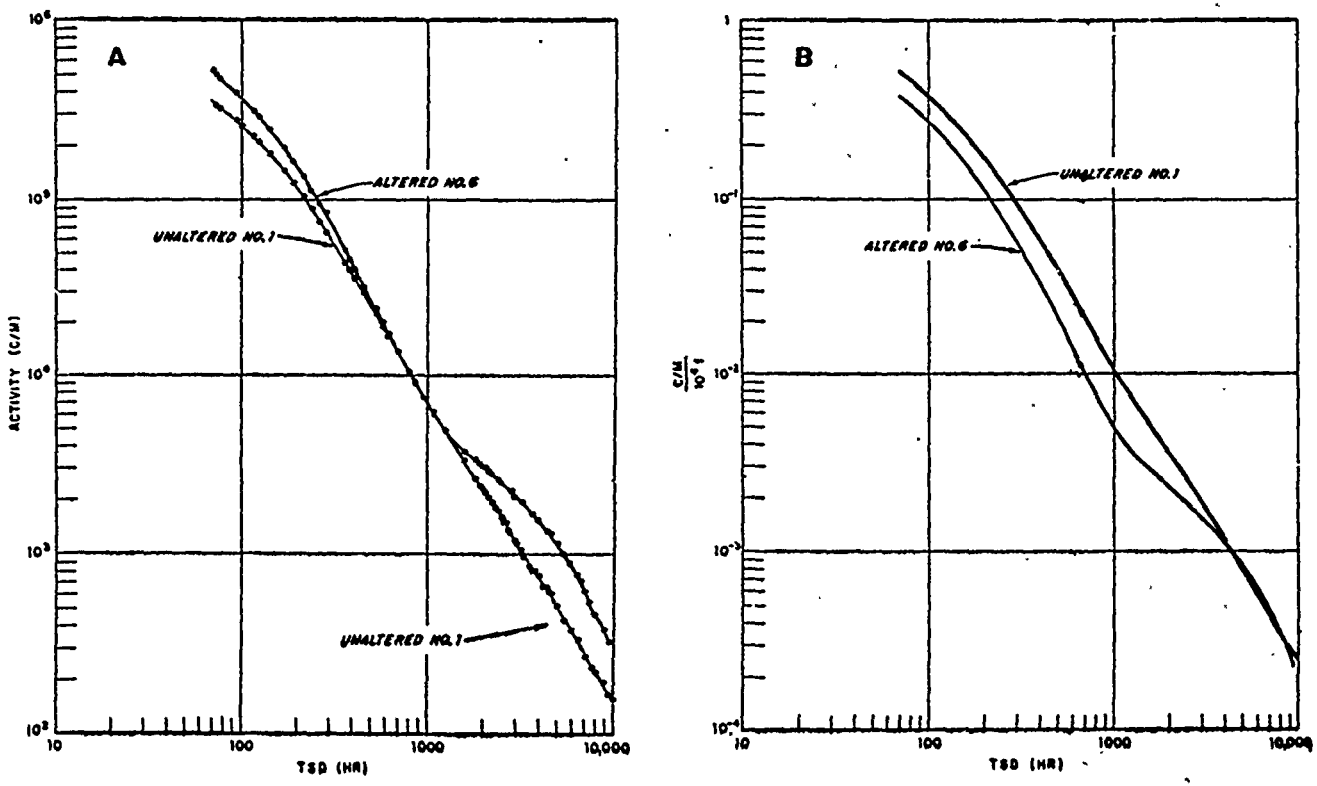

Fig. 5 WC Decay of Altered and Unaltered Particles Versus Time
A. Observed Activity.
B. Observed Activity per 10^4 Fissions.

Gamma ray spectra

In addition to rates of decay, the effects of differences in nuclide composition are shown by comparison of gamma spectra of altered and unaltered particles taken at various times after detonation. These spectra are shown in Fig. 6. Although differences are readily observed, the complex pulse height distributions are not easily resolved without extensive radiochemical data.[34] For example, in Fig. 6 the 0.1-Mev photons from Np^{239} are considerably more abundant in particle No. 6 than in No. 1. The result is a more rapid decay at early times for particle No. 6 as shown earlier in Fig. 5A. The reasons for differences in decay rates at about H+2000 hr, however, are not readily apparent on the basis of the observed gamma spectra shown in Fig. 6.

Additional differences are shown by comparison of the total fission values for the samples in Fig. 6 (Particles No. 151, 152, 170 and 177) with the gamma spectra above 0.4-Mev. These spectra clearly indicate a relatively higher abundance of photons above this energy in particles of unaltered composition. This is in good agreement with measured R values for the 140 mass chain which were over an order of magnitude higher in the unaltered than in the altered particles.

Fissions per gram of fallout

Table 4 reports mean fissions per gram of fallout for those samples which were weighed and analyzed for total fissions. Using the activity ratios (WC cpm/10^4f, ma/10^4f) given in Table 4 it was possible to compute the total fissions for a number of samples which had been weighed and counted but not analyzed for total fissions. The results of such calculations are given in Table 5, which also includes all samples which were analyzed for total fissions except for the combined sample WI.* The particles have been grouped according to class and the calculations made on the basis of activity ratios for each class as given in Table 4. Average values were used for those samples which had been measured in both the WC and GIC. The mean values for fissions/gram as given in Table 5 differ by approximately a factor of 100. This difference when compared with the data given earlier in Table 2 indicates that a very significant fraction of the total activity in the WHIM sample was from particles of the altered category. The latter observation is in agreement with the individual WHIM samplings presented in Table 3 and Fig. 2 which show that approximately 80 % of the well counter activity at H+382 hr was from the altered samples and an even higher percentage of the total fissions.

*The fission/gram value for this sample, 0.39 x 10^4, yielded a deviation from the mean of approximately seven times the probable error of the mean and was not used in the computations.

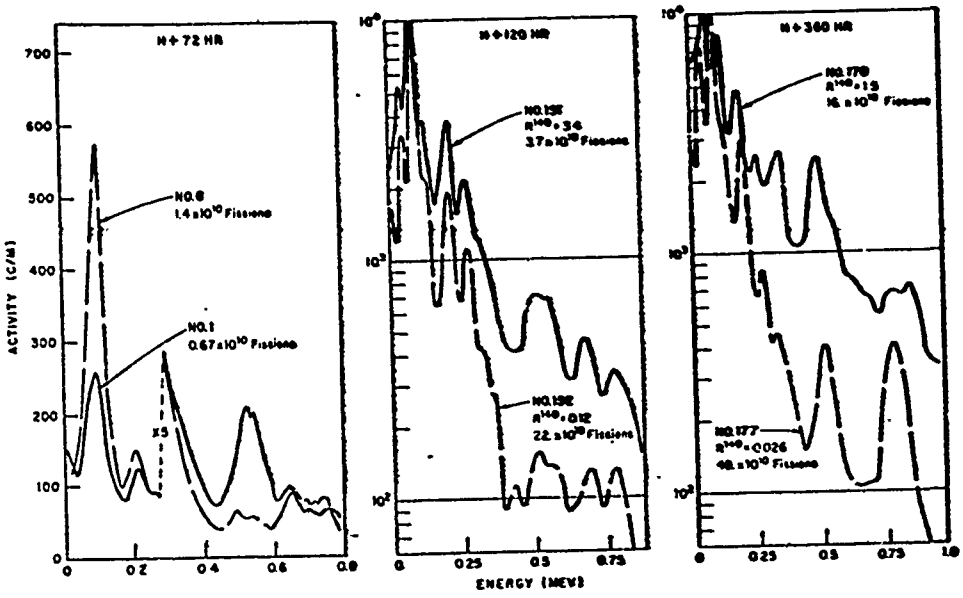

Fig. 6 Gamma Pulse Height Distributions of Altered (Nos. 6, 152, 177) and Unaltered (Nos. 1, 151, 170) Particles.

TABLE 5

Fissions/Gram Values for Altered and Unaltered Particles

Source	Lab. No.	Fissions x 10^4 per gram
	Altered	
YAG-40	2	1.2
OCC	152	1.0
	159	3.3
	163	0.51
WHIM (prior to sieving)	165W	4.7
	166W	4.8
	168W	8.5
	174	6.1
	175	6.3
	177	2.8
	179	2.6
	181	2.0
WHIM (500-1000 μ)	YS	6.7
	WS	8.2
	MEAN	4.2 ± 2.7
	Unaltered	
OCC	148	0.039
	149	0.027
	150	0.022
	151	0.045
	153	0.0010
	155	0.081
	156	0.024
	158	0.018
	160	0.070
	161	0.011
	162	0.10
	164	0.032
	165	0.016

TABLE 5 (Cont'd)

Fissions/Gram Values for Altered and Unaltered Particles

Source	Lab. No.	Fissions x 10^4 per gram
	Unaltered (Cont'd)	
OCC	166	0.0033
	167	0.031
	168	0.019
WHIM (prior to sieving)	167W	0.041
	169	0.0071
	170	0.033
	173	0.15
	176	0.0064
	178	0.0031
	180	0.0026
	182	0.0066
	MEAN	0.033 ± 0.035

R values

With respect to fractionation of radionuclides it has long been accepted that the mass 89 and mass 140 chains which exist for long time periods as noble gases, halogens and alkali metals* would condense late and therefore disproportionate with respect to less volatile elements. On the basis of long-lived gaseous precursors it would be predicted that the altered or melted particles would exhibit low R values for both chains, with the 89 smaller of the two. This was verified by the mean R values given in Table 4, which were 0.090 and 0.018 for the 140 and 89 chains, respectively. The corresponding values for the unaltered particles of 2.1 and 0.65 indicate that this latter class of particles may be important as a scavenger of these nuclides.

It is also of interest to compare R values obtained in this study with values obtained on gross fallout samples. The latter data gave Ba^{140} R values and Sr^{89} R values of 0.10 and 0.04 respectively** in the lagoon samples. The low R values for the gross sample from the lagoon area are similar to R values obtained with altered particles and suggests a lagoon fallout composed primarily of altered particles. This suggestion is supported by the WHIM sample fission/gram data (described above).

* Ba^{140} is formed by the decay of the radioelements Xe^{140} (16-sec half-life) and Cs^{140} (66-sec half-life); Sr^{89} is formed by the decay of the radioelements Kr^{89} (3.16-min half-life) and Rb^{89} (15.4-min half-life).

** P.D. LaRiviere, USNRDL, personal communication.

VALIDITY OF THE DATA

The data presented in this report are of three distinct types. These are weight determinations; activity measurements (in terms of instrument response, e.g., milliamps, counts); and radiochemical determinations for total fissions and R values.

The weight determinations were carried out with an accuracy and precision of approximately plus or minus 5 micrograms. Thus, the majority of the weight determinations were made to better than $\pm$ 1 %.

With few exceptions all determinations of activity by gamma well counting (WC) were carried out with a statistical precision of $\pm$ 3 % or better. The ionization chamber measurements (GIC) were made with a precision dependent upon the total activity. For samples reading greater than 10^{-9} ma the precision was better than $\pm$ 5 % and decreased from this value to approximately $\pm$ 20 % for samples which gave readings of 2x background (17×10^{-11} ma).

Experience with large numbers of samples has shown that the precision of the various radiochemical methods used in this study varied from $\pm$ 10 to $\pm$ 15 %. Since measurements of total activity were usually made with a far greater precision than the radiochemical analyses, the errors in the computations of the various activity ratios in terms of fissions were almost entirely dependent on the errors in radiochemical analysis. The accuracy of the radiochemical determinations were generally estimated as correct to $\pm$ 25 % but may have been as low as + 50 % for samples of low activity.

However, it is an unfortunate circumstance of experiments of this type that a large number of variables, many uncontrolled or unknown, exist which could introduce errors greater than those present in the methods of analysis. It is also generally true that little is known concerning the magnitude of such errors. Among errors of this kind may be listed the following:

1. Alteration of sample by collector, in transit, or in sampling process.
2. Non-random or biased selection.
3. Assumptions concerning reference standards.

Concerning the third point it is customary to base calculations for total fissions on the measured number of atoms of the reference nuclide, Mo^{99}. Although comparisons made on the basis of the latter quantity alone are valid it is apparent that estimates for total fissions or fractionation behavior of additional isotopes are actually based on the distribution of the reference nuclide throughout the fallout sample.

Although it was not possible on the basis of the available data to assess the magnitude of errors 1 and 2 it was possible to test the consistency of individual particle data with data from measurements on gross samples of fallout. The latter samples are less subject to the above errors than are individual particle samplings, are well-defined, and are analyzed with reasonably well-defined precision and accuracy.[29,35]

Comparison calculations were carried out with data obtained from particles from the OCC collector, How-F-67. The data for these particles have been given in Table 1. Following their selection from the OCC tray the particles were spread over a new tray and were counted as a group in a low geometry scintillation counter. Since the original tray had been previously assayed in the same counter and its total fission content determined,[36] it was possible to calculate the total fissions for the group of single particles as follows:

1. Total fissions, original tray (OCC How-F-67) = 1.78×10^6 cpm at H + 100 = 4.3×10^{14} fissions.
2. Activity, particles 148 to 168 = 6.02×10^3 cpm at H + 120 = 7.16×10^3 cpm at H + 100.
3. Total fissions, particles 148 to 168 = $(4.3 \times 10^{14})(7.16 \times 10^3 / 1.78 \times 10^6) = 1.7 \times 10^{12}$.

The value obtained, 1.7×10^{12} fissions, compares favorably with the total, 1.4×10^{12} fissions, derived by summing the number of fissions per particle as determined by direct radiochemical measurement or by calculation using the WC and GIC activity ratios given earlier in Table 4. A summary of fission values for each particle leading to the 1.4×10^{12} figure is given in Table 6.

TABLE 6

Summary of Individually Measured or Computed Fissions for Particles from OCC Tray How-F-67

Particle Number	Fissions Computed on basis of WC Measurements and activity ratio	Fissions Computed on basis of GIC Measurements and Activity Ratio	Fissions Measured by Mo^{99} Analysis	Measured or Average Computed Total (10^{10} Fissions)
148			6.4	6.4
149	1.55	1.68		1.6
150			1.9	1.9
151			3.7	3.7
152			22.	22.
153	.0328			.033
154	.0192			.019
155			2.6	2.6
156			1.0	1.0
157	3.34	4.50		3.9
158	0.0311			0.031
159			75.	75.
160			0.64	0.64
161	0.189	0.220		0.20
162			0.87	0.87
163			17.	17.
164	0.104	0.147		0.13
165	0.100	0.110		0.11
166	0.0857	0.0917		0.089
167	0.436	0.587		0.51
168	0.188	0.239		0.21
			SUM	138.

CONCLUSIONS AND RECOMMENDATIONS

Since the reported analyses were carried out on particles from a single detonation, a primary conclusion is that the analysis of individual particles created by certain types of detonations is entirely feasible. This feasibility was established since individual particles which represented approximately 10^{19} or more fissions were obtained. Thus it was possible to make quantitative measurements of the radioactivity of such particles.

Physical inspection of the fallout particles revealed several types of particles, each of which usually could be classified into one of two general divisions. The first particle class was made up of spherical or flaky particles. These were designated altered particles inasmuch as they represented an obvious physical alteration of normal coral particles. The second class consisted of angular or irregular particles and were designated unaltered particles. The most significant differences in the two classes were found to be in the amount and nature of the radioactivity associated with the particles. Based on the assumptions of (a) a radionuclide composition dependent only on particle class and not time or point of collection and (b) no serious disproportionation of the reference nuclide, Mo^{99}, by particle class it was found that the two particle categories differed in several respects among which were the following:

1. On the basis of simple arithmetic means the altered particles were approximately 100 times higher in terms of fissions per gram of total mass than were the unaltered.
2. The R values for Ba^{140} and Sr^{89} were over an order of magnitude lower in the altered particles than in the unaltered particles.
3. Inspection of Ba^{140} R values and comparison of total fission values with gamma spectra above 0.4 Mev, indicated a relatively higher abundance of photons above this energy in particles of unaltered composition.
4. In a sample collected near the site of detonation a very high percentage of the total fissions were due to particles of the altered category.
5. Decay rates for the altered and unaltered particles differed markedly at times less than H+100 and greater than H+1000 hours.

Some discussion and possible explanations for the observed data has been given earlier. As a result, it is suggested that fallout data obtained on gross particulate samples collected at various distances from the site of detonation, where possible, should be examined on the assumption of sample compositions consisting of varying percentages of altered and unaltered fallout particles as defined in this report.* That is, the data should be inspected to determine if fractionation effects, differences in gamma energy flux per fissions, etc. may be attributed to the relative numbers of altered and unaltered particles. Presentations of single particle data in terms of fissions/gram, R values, rates of decay, gamma spectra and activity ratios should aid in the interpretation of gross sample values. The individual particle information may also be of value in fallout model derivations and predictions and in particle formation theory.37

Further individual particle analyses should place greater emphasis on the collection and preservation of undisturbed samples and more rapid and improved methods of measurement. For particles of sufficient activity it was found that radionuclide differences were readily detected by measurement in two instruments with different gamma ray efficiencies. The gamma ionization chamber in conjunction with the well scintillation counter served satisfactorily in this respect except for the rather low sensitivity of the ionization chamber. With the exception of the gamma ray spectrometer measurements for total fissions the radiochemical separations were too slow for large numbers of samples. Either more rapid methods of separation and counting are required or partial separations followed by intensive gamma ray spectrometer measurements.

Approved by:

E. R. Tompkins

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For the Scientific Director

*Preliminary calculations carried out by P. LaRiviere, USNRDL, and the authors indicate that R values for gross samples are calculable from single particle radiochemical data and knowledge concerning the relative numbers of altered and unaltered particles.

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